

Relevance of intra- and inter-subband scattering on the absorption in heterostructures

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We analyze the absorption lineshape for inter-subband transitions in disordered quasi 2D heterostructures by an exact calculation. The intra-subband scatterings control the central peak while the tails of the absorption line are dominated by the inter-subband scattering terms. Our numerical study quantitatively assesses the magnitude of the free carrier absorption. The accuracy of different models currently used for gain/absorption is discussed.

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The demand to produce reliable terahertz and infrared devices has triggered extensive research on intersubband transitions in semiconductor heterostructures¹. A highlight is the realization and continuous improvement of Quantum Cascade Lasers (QCL)²⁻⁵ that emit in this frequency range. The search for improved performances of QCLs includes a better design of the layer sequence to enhance the population inversion between the two subbands involved in the lasing transition, the decrease of the non radiative paths, but also the control of the photon re-absorption by the free carriers that are present in the structure⁶⁻¹². The free carrier absorption (FCA), well known from bulk structures, is transferred to a variety of intra-subband and inter-subband oblique (in 2D \vec{k} space) transitions in heterostructures^{13,14}. Tailoring these (often parasitic) processes requires a thorough understanding of the nature of the optical transitions in imperfect heterostructures.

For light polarized along the growth (z) axis of an ideal semiconductor heterostructure, the energy dependence of the absorption coefficient between subbands 1 and 2 is a Dirac delta function line centered at $\hbar\omega_0 = E_2 - E_1$ ¹⁵. This “atomic-like” profile stems from the fact that the electromagnetic wave coupling requires a non-vanishing z -dipole $\langle 1|\hat{p}_z|2\rangle$, but does not affect the in-plane motion of the electrons. These motions are identical in the two subbands (i.e. plane waves, owing to the in-plane translation invariance) and their corresponding energies form identical dispersions (if we assume the same parabolic mass m^*). Static disorder affects the in-plane motion by introducing intra-subband ($V_{\text{def}}^{\text{intra}}$) as well as inter-subband ($V_{\text{def}}^{\text{inter}}$) couplings. Bound states usually appear below the 2D subband edges. As a consequence, the strict selection rules leading to the Dirac lineshape are no longer expected to apply to a disordered structure which is frequently taking into account by some effective broadening. Here we treat this feature from a fully microscopic point of view by considering the exact eigenstates of the disordered quantum well potential. Our

calculations allow us to gain a complete insight into the underlying physical processes leading to the absorption in heterostructures, not only near resonance, but also far above or far below the resonance (these regions have not deserved enough attention in the literature, even though of great technological interest). We will show that the regions far from resonance can nicely be understood by recurring to defect-induced scattering processes, and highlight the respective roles of $V_{\text{def}}^{\text{intra}}$ and $V_{\text{def}}^{\text{inter}}$. Finally, the complete calculation will allow us to determine the accuracy of different models currently employed to evaluate the gain/absorption of such structures.

In the presence of disorder, the heterostructure eigenproblem can be written as $\Psi_\nu(\vec{\rho}, z) = \sum_n F_{\nu,n}(\vec{\rho})\chi_n(z)$, with:

$$\begin{aligned} \frac{-\hbar^2}{2m^*}\nabla^2 F_{\nu,n}(\vec{\rho}) + \sum_{n'} F_{\nu,n'}(\vec{\rho})V_{\text{def}}^{n,n'}(\vec{\rho}) \\ = (\varepsilon_\nu - E_n) F_{\nu,n}(\vec{\rho}) \end{aligned} \quad (1)$$

where $\vec{\rho}=(x,y)$, $F_{\nu,n}(\vec{\rho})$ is the in-plane envelope function of the n^{th} subband and $V_{\text{def}}^{n,n'}(\vec{\rho}) = \langle \chi_n(z)|V_{\text{def}}(\vec{\rho}, z)|\chi_{n'}(z)\rangle$. $\chi_n(z)$ is the heterostructure localized part and E_n the corresponding bound state. In absence of defects $\nu \rightarrow (n, \vec{k})$, $F_\nu(\vec{\rho}) \rightarrow \exp(i\vec{k} \cdot \vec{\rho})/\sqrt{S}$ and $\varepsilon_\nu \rightarrow E_n + \hbar^2 k^2/(2m^*)$. In the following we shall present results obtained after a numerical diagonalization of Eq. (1) within a truncated basis with the two lowest subband states ($n=1,2$). In practice, we expand $F_{\nu,n=1,2}(\vec{\rho})$ in a plane wave basis that fulfill periodic conditions in a 200×200 nm² box. The matrix element of the dipole coupling to light are proportional to $\langle \hat{p}_z \rangle_{(\nu,\mu)} = \langle \Psi_\nu(\vec{r})|\hat{p}_z|\Psi_\mu(\vec{r})\rangle$. For defect-free structures one has the selection rules: $\langle \hat{p}_z \rangle_{(n,\vec{k}), (n',\vec{k}')} = (1 - \delta_{n,n'})\delta_{\vec{k},\vec{k}'}\langle \chi_n|\hat{p}_z|\chi_{n'}\rangle$, reflecting the in-plane translation invariance and the non-vanishing z -dipole. In the following, we will be interested in the effect of disorder on these matrix elements.

In QCLs interface defects constitute an important elas-

tic scattering mechanisms, which we will consider in detail here. Most features can be directly transferred to other mechanisms such as impurity and alloy scattering. Interface defects arise from the non ideality of the well/barrier interface at $z=z_0$ between two consecutive layers of the heterostructure: they can be a protrusion of the barrier material in the well (repulsive defects) or vice-versa (attractive defects). For nearly ideal interfaces, the interface defects are only one monolayer thick. The scattering potential created by one disordered interface has the form

$$V_{\text{def}}(\vec{r}) = f(z)u_{\text{def}}(\vec{\rho}) \quad (2)$$

showing a separation on the dependence on the z and $\vec{\rho}$ variables. $u_{\text{def}}(\vec{\rho})$ is the superposition of localized functions centered at the randomly placed interface defects on the (x, y) plane. For a barrier/well interface at $z = z_0$ and for a repulsive defect $f^{\text{rep}}(z) = V_b\Theta(z - z_0)\Theta(z_0 + h - z)$ while for an attractive defect $f^{\text{att}}(z) = -V_b\Theta(z_0 - z)\Theta(z - z_0 + h)$, where V_b is the barrier height, h is the thickness of one monolayer, $\Theta(z)$ is the Heaviside function.

In Fig. 1 we show the matrix $|\langle\Psi_\nu(\vec{r})|\hat{p}_z|\Psi_\mu(\vec{r})\rangle|^2$ for our calculated eigenstates of the disordered heterostructure. Here we consider a 9/2/3 nm GaAs/Ga_{0.75}Al_{0.25}As double quantum well (DQW) structure with Gaussian interface defects placed on the two inner interfaces, with a characteristic in-plane extension of 3.6 nm and a fractional coverage of the surface $N_{\text{def}}\pi\sigma^2/S = 30\%$. The figure clearly displays two blurred straight regions around $|\varepsilon_\mu - \varepsilon_\nu| \approx \hbar\omega_0 = 73.8$ meV, corresponding to the subband spacing in this sample. If there were no disorder, there would be no blurring since (see above) a single final state would match any given initial state. The fact that, the matrix element is almost zero if the energy difference between the true states differs strongly from the inter-subband spacing corresponds well with the conventional broadening picture.

In this context it has to be noted, that the blurring is strongly reduced, if the wave functions $\chi_n(z)$ for the subbands $n = 1$ and $n = 2$ are of the same magnitude at each interface. Neglecting $V_{\text{def}}^{\text{inter}}$ this provides identical (or very similar) in-plane wave functions for both subbands, which provide strong selection rules for the z -matrix elements. Similar selection rules appear in strong magnetic fields¹⁶. In our case, the wave functions differ essentially at the interfaces, as the excited state penetrates stronger into the barrier, so that this effect is only of very minor relevance.

We show in Fig. 2 (red full line) the absorption coefficient of the DQW structure based on the matrix elements of Fig. 1 (in all absorption calculations, the delta of energy conservation was replaced by a Gaussian with a FWHM=1.88 meV). In this absorption spectrum, the contributions of $V_{\text{def}}^{\text{intra}}$ and $V_{\text{def}}^{\text{inter}}$ are taken at all orders and are fully admixed. In order to highlight the physics underlying the assisted light absorption and disentangle the various contributions to its spectrum, we

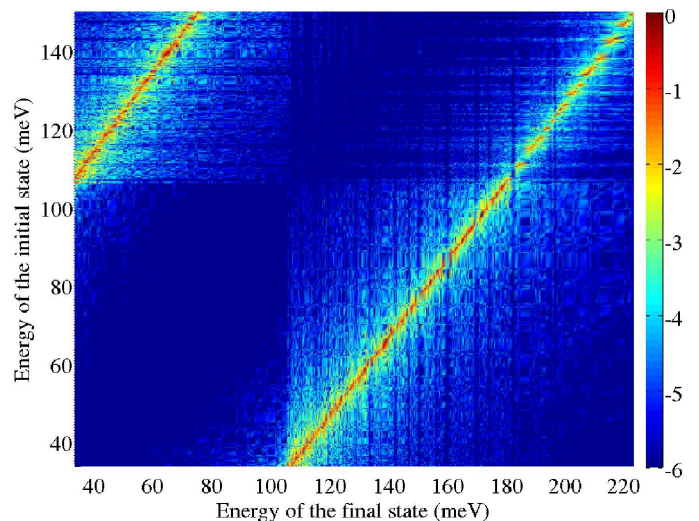


FIG. 1. Color plot of the decimal logarithm of the normalized squared modulus of the optical matrix elements for the ensemble of transitions $\varepsilon_\nu - \varepsilon_\mu$ in a disordered 9/2/3 nm GaAs/Ga_{0.75}Al_{0.25}As double quantum well. The disorder is due to interface defects randomly distributed on the two inner interfaces of the structure (see text).

discuss in the following the results obtained within different approximations and/or models. We also show in Fig. 2 the calculated absorptions when only $V_{\text{def}}^{\text{intra}}$ (green dotted line) or only $V_{\text{def}}^{\text{inter}}$ (orange dashed-dotted line) are retained in the scattering matrix. We see that the low energy (the high energy) FCA is dominated by the $V_{\text{def}}^{\text{inter}}$ ($V_{\text{def}}^{\text{intra}}$) couplings. Indeed, the initial and final states involved in the non-resonant absorption displays a dominant E_1 or E_2 subband character. Thus, since the dipole coupling only triggers inter-subband events and starting from an initial state in E_1 , an additional inter-subband scattering is needed for the low energy intra- E_1 oblique transition. Conversely, the high-energy absorption tail involves essentially E_1 -to- E_2 processes and thus rely mostly on $V_{\text{def}}^{\text{intra}}$. It is worth pointing out that the near-resonant absorption (see Fig. 2b) is accurately rendered by the only-intra approximation, whereas the only-inter one fails in predicting both the intensity and the absorption profile. Finally, we also show in Fig. 2 the perturbative estimates of the FCA obtained by expanding the electron wavefunction to the first order in V_{def} ¹⁴ (dashed blue line). We see that the perturbative estimate provides an excellent rendering of the shape and strength of the actual absorption when it is justified, i. e. not too close to the resonance.

An important issue is to understand how the results of the exact diagonalization compare to correlation function approaches, essentially based on Green's function methods. To this end, we show in Fig. 3 the absorption coefficient for the $E_1 - E_2$ transition of the DQW structure described above, calculated with two different correlation function (CRF) approaches, and we compare it with

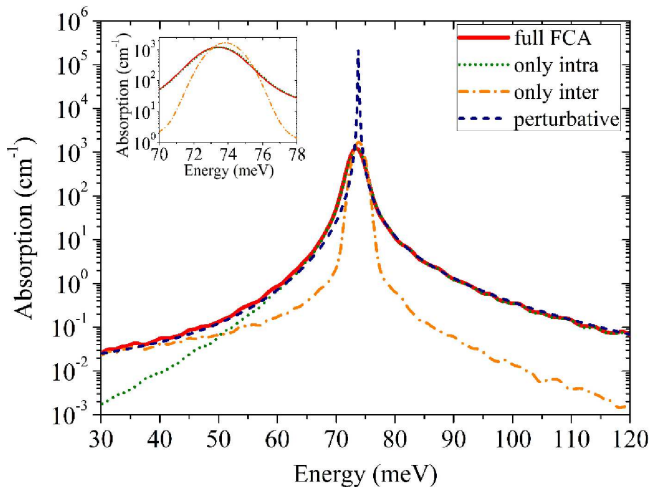


FIG. 2. Absorption spectrum for the E_1 - E_2 transition calculated by: fully numerical diagonalization (red full line); taking into account either only $V_{\text{def}}^{\text{intra}}$ (green dotted line) or only $V_{\text{def}}^{\text{inter}}$ (orange dashed-dotted line); and by expanding the electron wavefunction to the first order in both $V_{\text{def}}^{\text{intra}}$ and $V_{\text{def}}^{\text{inter}}$ ¹⁴ (blue dashed line). Inset: Zoom of the absorption spectrum in (a) around $\hbar\omega_0$. $T = 100$ K.

the one calculated by exact diagonalization. In CRF_1 the Keldysh Green's function formalism was applied using the implementation of Refs. 17 and 18. In order to make a quantitatively meaningful comparison among the various calculations, we numerically extracted the correlation length and the average defect depth from the randomly generated interface defects used in the exact calculation without the use of any fitting parameters. We see that CRF_1 gives a good description of the low ω behavior, as the inclusion of the nondiagonal self-energies¹⁷ fully covers the inter-subband terms addressed above. At large energy, CRF_1 overestimates the absorption; a feature that is related to the use of k -independent scattering matrix elements in order to simplify this numerics. This assumption of an effective delta- potential scattering potential overestimates the scattering for large wave-vector transfer, which is crucial for the tails. For the peak, however, this approximation turns out to provide a full quantitative description, without any adjustable parameters. In CRF_2 , we use *Unuma et al.*¹⁹ formalism for the intersubband absorption coefficient that follows Ando's approach²⁰. To draw CRF_2 we converted *Unuma et al.*'s real part of the conductivity into an absorption coefficient following¹⁴ and we used the interface disorder potential described above. It is clear that CRF_2 poorly describes the far from resonance FCA. This is most probably due to the fact that, contrarily to CRF_1 , the off-diagonal components of the Green's function are neglected, whereas the diagonal ones are evaluated within the self-consistent Born approximation.

In conclusion, we have computed numerically the line-shape of inter-subband transitions in heterostructures with interface disorder. We have found that only few fi-

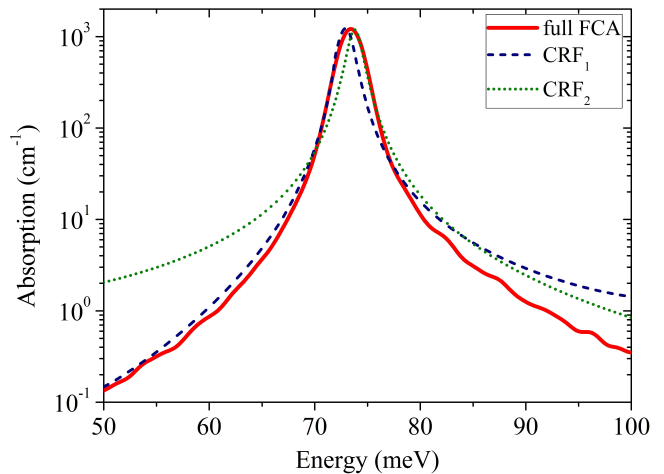


FIG. 3. Absorption spectrum for the E_1 - E_2 transition calculated by three different models: exact diagonalization (red full line), Keldysh Green's function formalism, CRF_1 ^{17,18} (blue dashed line), Unuma's model, CRF_2 ¹⁹ (green dotted line). $T = 100$ K.

nal states are optically connected to a given initial state despite disorder, which reflects the common picture of a broadened transition. While intrasubband scattering dominates at the resonance peak and at higher photon energies, intersubband scattering dominates the low energy tail of the absorption spectrum. We have compared the outputs of several models to compute absorption to the fully numerical results, in particular their low ω behaviors. The perturbative estimate for FCA¹⁴ works very well for the absorption tails. We find also that *Unuma et al.*'s model¹⁹ gives a poor description of the non-resonant FCA tails, while the Keldysh Green's function formalism¹⁷ with full nondiagonal self-energies fits the numerical results nicely. A clear picture of the inter-subband and intra-subband transitions now emerges from this study and emphasizes the need for a proper account of the perturbation of the carrier wavefunctions by defects and not only of their energies.

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